Environmental Science & Technology

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Mercury Emissions from Biomass Burning in China

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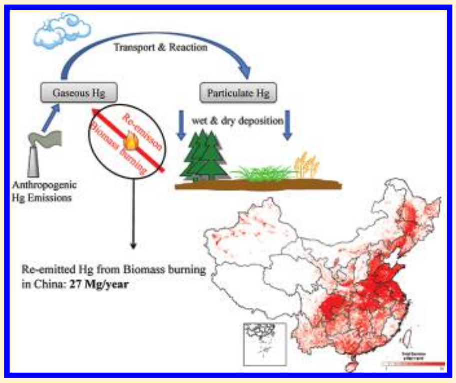
S Supporting Information

ABSTRACT: Biomass burning covers open fires (forest and grassland fires, crop residue burning in fields, etc.) and biofuel combustion (crop residues and wood, etc., used as fuel). As a large agricultural country, China may produce large quantities of mercury emissions from biomass burning. A new mercury emission inventory in China is needed because previous studies reflected outdated biomass burning with coarse resolution. Moreover, these studies often adopted the emission factors (mass of emitted species per mass of biomass burned) measured in North America. In this study, the mercury emissions from biomass burning in China (excluding small islands in the South China Sea) were estimated, using recently measured mercury concentrations in various biomes in China as emission factors. Emissions from crop residues and fuelwood were estimated based on annual reports distributed by provincial government. Emissions from forest and grassland fires were calculated by combining moderate resolution imaging spectroradiometer (MODIS) burned area product with combustion efficiency (ratio of fuel consumption to total available fuels) considering fuel moisture. The average annual emission from biomass burning was 27 (range from 15.1 to 39.9) Mg/year. This inventory has high spatial resolution (1 km) and covers a long period (2000−2007), making it useful for air quality modeling.



1. INTRODUCTION

Mercury is a toxic and persistent environmental pollutant. Atmospheric mercury is deposited in various pathways into the ground and water. Some of the mercury is transformed into methyl mercury, which bioaccumulates and biomagnifies in food webs, resulting in increased concentrations in higher organisms. Mercury remains an important subject of global pollution control efforts because of its toxicity and its involvement in the atmosphere-biosphere biogeochemical cycle and long-range transport.

Mercury emissions from biomass burning have recently received increasing attention due to their potentially significant contribution to the atmospheric mercury budget and particularly their impact on the global mercury cycle.[1] Earlier studies have estimated that the average global annual mercury emitted from biomass burning for 1997−2006 was 675 ± 240 Mg/year. This is equivalent to 8% of all currently known anthropogenic and natural mercury emission sources for the same period.[2] During the biomass burning process, mercury can be remobilized and re-emitted into the air. Biomass burning therefore accelerates emission and deposition cycles of mercury between terrestrial ecosystems and the atmosphere.

China is rich in mercury mineral resources; moreover, as a developing country, the amount of coal consumed in China approaches approximately 28% of the world's total consumption. Previous work has shown that atmospheric mercury emissions from nonferrous metal smelting and coal combustion in China could be the highest in the world.[3] However, as a large agricultural country, the mercury emissions from biomass burning in China could also be significant. The majority of the population lives in rural areas and biofuel (including crop residues, fuelwood, and animal dung) is an important energy source. Moreover, open burning, such as forest fires, grassland fires, and burning of field crop residues in rural areas also release mercury.[4]

A few attempts have been made to estimate the mercury emission from biomass burning to the atmosphere in China. Streets et al. estimated that 19 Mg of mercury was released from biomass burning in China during 1999, including grassland burning, forest burning, biofuel combustion and agricultural residues burning.[4] This inventory was developed on the basis of a variety of statistical data for the 1950s-1990s. The data employed in that work are outdated, as the burning activities in China have changed significantly during the last two decades, especially forest fires. Friedli et al. have evaluated that 7 ± 2 Mg mercury was released yearly from biomass burning other than biofuel combustion in Central Asia during 1997−2006.[2] Another limitation is that the emission factors adopted in these two studies were measured in North America, which might be inappropriate for China. China is known for high levels of atmospheric mercury from coal combustion and nonferrous metals smelting, which results in high deposition of

Received: June 29, 2011
Accepted: September 27, 2011
Revised: September 21, 2011
Published: September 27, 2011

mercury into the local terrestrial ecosystem and absorption by vegetation.[5] Third, both the results have 1° × 1° resolution, which could be too coarse for air quality modeling.

A new mercury emission inventory with fine resolution to reflect recent biomass burning emission in China is needed, especially for atmospheric simulation. In this study, the mercury emissions from biomass burning in China (excluding small islands in the South China Sea) during 2000−2007 were estimated. We adopted the newly available MODIS burned area product (MCD45A1) at a 500 m resolution to calculate the emissions from forest and grassland fires. Because the burned area products from remote sensors with medium resolution often miss the crop burning in fields due to its small size, we used the official statistics data at the provincial level to estimate the mercury emissions from crop residues burning in fields and biofuel combustion in homes. The mercury concentrations for different parts of biomass (leaf and stem) measured in recent studies in China were used as surrogates for emission factors.[6−8] Finally, the inventory is allocated spatially at 1 km resolution by using the Global Land Cover Data set 2000 (GLC-2000), MODIS thermal anomalies/fire products (MOD14A2 and MYD14A2) and rural population density.

2. METHODOLOGY AND DATA SOURCE

Emission Factors. Mercury emission was estimated using the product of the amount of biomass burned and emission factors (EFs). Previous researches adopted the EF values measured in North America to estimate the mercury emissions in China because native measurements of EFs in China were not available.

Several studies showed that mercury contained in vegetation (live, dead, coniferous, and deciduous) was essentially completely released in burns. The speciation of the emitted mercury is primarily in the form of gaseous elementary mercury (GEM) for both dry and green fuels.[6,9] Mercury concentrations in vegetation and soil are treated as the EFs in some estimates of mercury emissions from biomass burning.[6−8] Fortunately, a few recent studies reported the mercury concentrations in various biomes in China. Because the majority of forest fires in China occur in the northeast regions,[10] mercury concentration measurements carried out in this area were chosen as EFs for Chinese forest fires. Since more crop residues were burned in south China,[11] we chose the experimental mercury concentration results from the southern part of the country as the EFs for crop residue burning. The biomass ratio of stems/leaf and EFs are averaged from the reported results, as listed in Table 1.

The emissions from stem and leaf burning should be calculated separately because the mercury content in different tissues may be very different. Mercury concentrations are much higher in leaf tissue than in the stems. This can be attributed to the fact that nearly all of the mercury in both herbaceous plants and woody plants was absorbed from the atmosphere by stomata on the surface of the foliage. This is the predominant pathway by which mercury accumulates in plants.[12]

Recent studies found concentrations of ambient mercury in China that are much higher than in other countries.[13] As shown in Table 1, EFs for biomass burning in China are greater than the values in other countries. Results from recent investigations of Hg concentrations across 14 forest site in the United States shows that average Hg concentrations in stems and foliage are 15 and 40 ng/g respectively.[14] The concentrations measured in China for forest leaves are, on average, 11% higher than the

Table 1. Emission Factors Used for Different Biomass Categories

biomass type	biomass of stem/leaf	EFs used in this study (ng/g)[b]		comparison of total mercury concentration (ng/g)[a]	
		stems	leaves	China	other countries
forest and fuelwood	5.0[35]	40 (17−76)[26]	100 (21−204)[26]	7−96(l),[36] 18−122(l), 6−68(s),[37] 31−45(l), 17−37(s),[38] 23−32(l),[39] 15−883(l), 6−233(s),[26] 21−35(l)[40]	19−133(l), 24−27(s),[9] 20,[6] 38−190,[41] 0−130,[42] 60,[1] 42−72,[6] 1−57(s),[14] 8−48(l)[14]
grass		24[40]	24[40]	24,[40] 34[38]	6,[2] 18−39[43]
crop residue	3.0[44]	35 (9−221)[5]	319 (84−1394)[5]	84−1394(l), 9−221(s),[5] 70−130(l), 11−21(s),[45] 30−54(s),[46] 1−22(s)[47]	7.8−9.8(s),[9] 37[41]

[a] L and s in parentheses refer to leaves and stems, respectively. [b] Values in parentheses are the ranges of EFs.

corresponding values in the United States and those for crop stems are 60−138% higher. In addition, approximately 3−7 times more mercury was found in China's grasslands than that in Africa's grasslands. The reason for this could be that Chinese mineral resources are rich in mercury, the control measures during coal burning, smelting and mining are poor, and atmospheric deposition is correspondingly high. Finally, the accumulated mercury is re-emitted into the atmosphere during biomass burning process.

Forest and Grassland Fires. Mercury resides both in the above-ground biomes and organic soil. Nevertheless, histosols is very sparse in China and its emission is relatively small and could be neglected.[15] The amounts of burned biomass from forests and grasslands were computed based on the 500 m MCD45A1 burned area product, fuel load and combustion efficiency.[16] The MCD45A1 product was validated for forest burning in China by comparing MCD45A1 results with the fire-affected forest area recorded by the National Forestry Bureau, by month and province from 2001 to 2006.[17] Fuel loads for individual provinces were assigned by vegetation types. Impacts of fuel type and fuel moisture were taken into account in calculating combustion efficiency.[18] More details can be found in Song et al.'s paper.[19]

In-field Burning and In-home Combustion of Crop Residues. Crop residues, including residues from rice, wheat, corn, coarse cereals, cotton, legumes, peanut, or rape, are often burned for household energy and used as fertilization in the field. The emissions from this burning were found to be significant.[20] Crop fires in the fields in China were often missed by MODIS due to their small size. In this study, the provincial amounts of crop residues burned in fields and in homes as fuel are estimated by multiplying the total crop residues, the percentage of field/domestic burning of crop residues and the combustion efficiency. The total crop residues is the product of crop productions at the provincial level distributed by the government and the residue/crop ratio (listed in Supporting Information (SI) Table S1).[11] The percentage of crop residues that were burned in fields or in homes as fuel were adopted from a large-scale investigation on the usage of crop residues in different provinces, which provides crop-specific percentages of domestic and field burning (see SI Table S2 and S3).[20] Combustion efficiencies (SI Table S1) are specified by crop type. Much more of the crop residues were used as fuel in densely populated rural areas; this is because these populations have less income and poor access to other energy sources. In the developed regions, the crop residues are more likely to be burned in the fields.

Fuelwood Combustion. As another important energy source, fuelwood is widely distributed and available in many remote mountainous areas of China. For instance, more than 10 Tg fuelwood is consumed every year in Guangxi and Hunan. In this study, the amount of fuelwood combustion at the provincial level was estimated on the basis of fuelwood consumption and combustion efficiency.[10] The combustion efficiency of fuelwood was assumed to be 87%.[21]

Spatial Allocation. The aim of this study is to produce a mercury emission inventory with fine resolution. Although the amount of crop residues burnt in fields in China could not be reflected accurately in burned area products (MCD45A1) because of their small size, they could be located by MODIS fire counts data.[22] We selected 2002−2007 MOD14A2 and MYD14A2 products (MODIS Thermal Anomalies/Fire 8-Day 1 km L3 global products from satellites Terra and Aqua, when Terra passes over China at 10:30 while Aqua is at 14:30 local time, respectively) to determine the open fire frequencies and spatial distribution on a 1 km grid.[23] Only open fires in the land cover classes defined as "Farm" and "Mosaic of cropping" in the GLC-2000 land cover data set (also at 1 km resolution) were identified as crops burning in fields. The mercury emission in the i-th zone (E_i) was calculated using the following equation:

$$E_i = \frac{FC_i}{FC_k} \times E_k$$

where FC_i is the fire count in i-th zone, FC_k is the total fire count in province k and E_k is the total estimated mercury emission from crop residues burning in fields in province k.

Similarly, the provincial level emissions from in-home combustion of crop residue and fuelwood combustion are allocated by using the rural population density in 1 km zones.[24] The forest and grass fires emission data were derived with 500 m spatial resolution. Finally, all the emissions were aggregated at 1 km resolution.

Table 2. Mercury Emissions (Mg) from Biomass Burning in China during 2000-2007

year	total	forest	grassland	field crop residues	crop residues as fuel	fuelwood
2000	24.2	0.01	0.00	4.3	14.2	5.7
2001	25.5	0.03	0.00	4.3	14.3	6.9
2002	26.9	0.02	0.00	4.2	14.7	8.0
2003	26.2	0.11	0.01	4.0	13.9	8.2
2004	28.4	0.08	0.01	4.4	15.4	8.5
2005	27.7	0.03	0.01	4.5	15.9	7.3
2006	28.4	0.03	0.00	4.6	16.1	7.6
2007	28.7	0.04	0.00	4.7	16.6	7.3
average	27.0	0.04	0.00	4.4	15.2	7.4
percentage	100%	0.1%	0.0%	16.2%	56.3%	27.4%

3. RESULTS AND DISCUSSION

Comparisons to Other Studies. The total annual mercury emissions from biomass burning in China during 2000−2007 are listed in Table 2, and range from 24.2 Mg (2000) to 28.7 Mg (2007). The average annual total mercury emission is 27.0 Mg.

The amount of burned biomass, combustion efficiency and EFs were the sources that caused the uncertainties in the estimation. Given the larger presumed uncertainties of statistics for untracked energy use, the probability of the value used for burned biomass amount was assumed to have a normal distribution with a coefficient of variation (CV) of 30%.[25] The typical uncertainty of the EF is 50%.[5,26] We ran 20000 Monte Carlo simulations to estimate the range of fire emissions with a 90% confidence interval. The estimated emission range is 15.1−39.9 Mg/year.

The emissions from crop residues combusted in homes is the biggest contributor to the total mercury emissions (56%), followed by fuelwood combustion (27%) and field burning of crop residues (16%). Emissions from forest fires are relatively small, and grassland fire emission are negligible. Previous studies also found that crop residues and fuelwood were the dominant gaseous pollutants out of all biomass burning in China.[27]

Streets et al. estimated the total mercury emission from biomass burning in 1999 as 19.2 Mg, somewhat lower than our result, 27.0 Mg.[3] The biggest disparity is in the biofuel emission estimates. We estimated 7.4 Mg of emissions came from fuelwood and 15.2 Mg came from crop residue, for a total of 22.5 Mg of mercury from biofuel sources. Street et al. estimated only 8.3 Mg of mercury was released from biofuel sources in 1999.[3] The two biofuel amounts (the sum of fuelwood and crop residues) were comparable: 413 Tg in Street et al.'s paper and 349 Tg (the sum of fuelwood and crop residues) in our research. The fuelwood amount in the year of 2000, 185 Tg, was close to the 177 Tg in 2000 reported by Yan et al.,[28] and the biofuel amount from crop residues, 164 Tg, was close to the 106 Tg reported by Zhang et al. in 2004.[29] Thus, the high EFs in our study are probably the reason for the disparity. We adopted the recent measurements in China, 40 ng/g for crop stems and 100 ng/g for leaves, while Streets et al used the EF value of only 20 ng/g.

The other disparities between the two studies were the emissions from forest fires and grass fires. The forest fire emission in our estimation was 0.04 Mg, much lower than the 2.8 Mg reported in Streets et al.'s research. Streets et al. adopted 113 ng/g as the emission factor, comparable to ours (Table 1); however, they used government records of the 1950s−1990s as the burned areas, whereas we chose to use the 2000−2007 MODIS burned area products. The recent forest fires in China were captured very well, as confirmed by comparing MCD45A1 results with the monthly, provincial fire-affected forest area recorded by the government from 2001 to 2006.[17] Recent study also confirmed that forest fire activities have decreased drastically over the last two decades due to law enforcement.[15] For grassland burning, Streets et al. estimated 4.17 Mg mercury emission, while the grass burning emission in our results was small enough to be neglected. That study used a higher EF for grass, 80 ng/g. Another reason for the disparity is that the assumed grass burned areas are larger in their study. They assumed a uniform burned fraction over all of China equal to that in Mongolia (3.0%), while we derived grass burned areas again from the MCD45A1 product. The recent records showed that only 0.6% of total grassland was burned in China during 2000−2008.[30] The mercury emissions both from forest and grassland fires may therefore be overestimated by Streets et al.

The field crop burning contributed 3.9 Mg mercury in Streets et al.'s paper, close to the 4.4 Mg in our results. There was a difference in EF values: Streets et al. used 37 ng/g, and we used 35 ng/g for stems and 319 ng/g for leaves. Streets et al. estimated the ratio of crop residues burned in the field with a single value of 17% for every kind of crops over the whole country; however, in our estimates, the provincial-level and crop-specific percentages used were based on surveys from 2000. From our method, an average of 6.6% of crop residues was burned in field.

China accounts for approximately 4% of the global mercury emissions from biomass burning, 675 Mg as estimated by Friedli et al.[2] In contrast to the larger contributors to mercury emissions, such as the United States with 44 Mg yearly,[8] the majority of the emissions from China were from crop residues combustion (72%, including burning in fields and home fuel use), whereas the corresponding percentage in the United States was only 3%, the rest is mostly caused by forest fires. The total atmospheric mercury contributed from biomass burning in China was only 5% of that from anthropogenic activities (536 Mg) mercury, most of which was emitted from coal combustion and nonferrous metals smelting.[3] However, as proposed previously, biomass burning could play an important role in mercury exchange between terrestrial ecosystems and the atmosphere.

Temporal Variations. The total mercury emission from biomass burning ranged from 24 Mg in 2000 to 29 Mg in 2007. The annual variations could be attributed largely to the crop residues combustion from in-field and in-home use. The amount of crop residues used for domestic burning depends on the rural population and crop residues yield. The reduction of straw-based forage and the rising price of fossil fuel in recent years also promoted the usage of crop residues. The amount of crop residues burned in fields dropped from 4.3 Mg in 2000 to 4.0 Mg in 2003, and increased in 2004−2006.

Fuelwood consumption, another important contributor to mercury emissions, increased rapidly during 2000−2004 and fell slightly in 2005−2007. This fall could be caused by the government vigorously advocating the use of renewable energy sources and energy-saving measures in rural areas in recent years. In addition, an increasingly strict regulatory system of forest protection played an important role in the control of fuelwood consumption.

Forest and grassland fires made only a very small contribution to the total amount of biomass burning in China. The forest and grassland fires correlate with both natural causes (lightning, low precipitation, high temperature, etc.) and human causes (land clearing for agriculture or habitation, etc.). The emissions in 2003 are by far the highest (115 kg), and the minimum emissions occurred in the year 2000 (6 kg). Correspondingly, these two years exhibit the greatest and the lowest amounts of burned areas in the MCD45A1 product. The large forest and grassland fires in 2003 may have been influenced by El Niño and an abnormal climate.[31]

Spatial Distribution. The year 2006 was selected to demonstrate the spatial patterns, which are shown in Figure 1 and SI Figures S1−S3. In general, there is a high emission zone located in central China, which includes Sichuan, Hubei, Henan, Shandong, Jiangsu, and Anhui Provinces (see SI Table S4). Higher mercury emission was caused by larger rural population and their reliance on crop residue combustion. These provinces accounts for 42% of the total mercury emissions in 2006. This region is an important agricultural zone in China. The amounts of crop residues in these provinces are high. Moreover, the populations are also high; dense population needs more energy to consume. However, the economic income in such rural areas is often low. The forest cover is often lower in the agricultural areas, with less wood to be used as biofuel, and thus, crop residues become their most important energy source (SI Figure S2). The straw from wheat, corn, rice, and cotton can be burned easily. The percentage of these four crop residues used as domestic biofuel is 59% in Sichuan, 53% in Hubei and 45% in Jiangsu, as examples, which is much higher than the average of 24% for China as a whole.[20] Compared with the other provinces, the portion of crop residues burned in the fields in central China was lower.

The Southwestern provinces also play a significant role in mercury emissions from biomass burning, especially Hunan, Guangxi, Guizhou and Yunnan Provinces. Total mercury deposition fluxes measured in Guizhou range from 336 to 2340 $g/km^2/y$, much higher than average value for East Asia ($<50\ g/km^2/y$).[32] The reason is the high mercury content of raw coal in this province and the relatively large amount of uncontrolled coal combustion. This area contributed 17% of the total mercury emissions from biomass burning. Most of the people there live in mountainous areas. Unlike the agricultural provinces, the crop residues amounts are often much lower. The rural people here depend on wood combustion for energy. Fortunately, the forest coverage is high, approximately 43%, which is 25% higher than

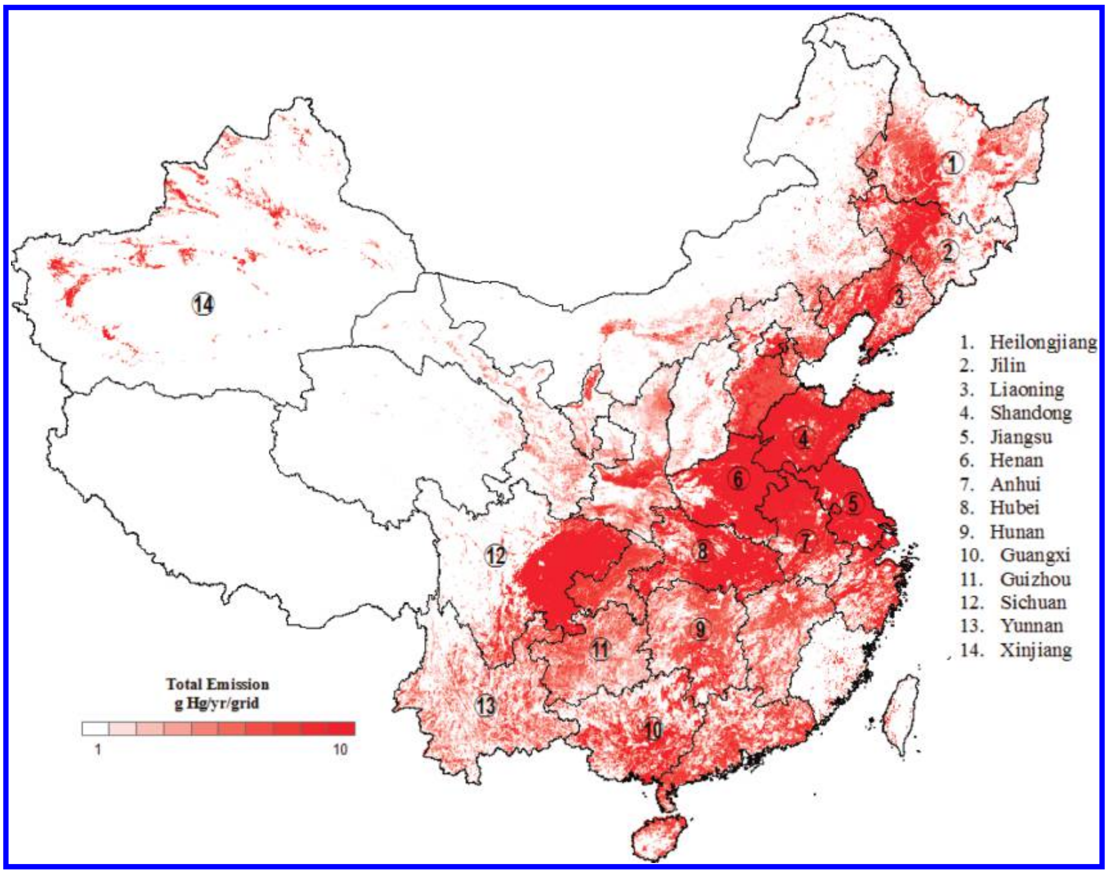


Figure 1. Mercury Emissions from Biomass Burning for the year 2006 (1 Km × 1 Km).

the average level for the whole country.[33] More than 50% of total mercury came from fuelwood combustion, as shown in SI Figure S2. In 2006, 12 Tg of fuelwood and 5 Tg of crop residues were burned as domestic fuel in Guangxi province.

Another important contributor is northeast China, including Liaoning, Jilin and Heilongjiang Province. The deposition flux measured in Jilin is approximately 320 g/km^2/y, which can be attributed to the presence of nonferrous metal smelters.[34] This area contributed 14% of the total mercury emission from biomass burning. As this area is an important producer of winter crops and has dense boreal forest areas, the amounts of emissions from fuelwood and crop consumption were also large. People need more energy for heating over the long winter. Moreover, it should be mentioned that the MCD45A1 product indicates that northern China was the largest contributor to the total burned forest areas (averaging 62% of the forest) during the period of 2000−2006.

The western region of China has the lowest emissions because of low population density and crop yields. However, high cotton and wheat yields in western Xinxiang lead to some higher local mercury emission from crop residue combustion.

In summary, China has a unique bioenergy consumption pattern which results in regional differences in mercury emissions. The mercury concentration in vegetation is very site-dependent and species-dependent. Since China is known for high levels of atmospheric mercury and correspondingly high deposition to vegetation, area specific measurement of Hg concentration values for various biomes in China are very desirable and necessary in future research. We also note that the mercury emission estimation from soil during open fires is needed.

ASSOCIATED CONTENT

S Supporting Information. Tables for crop-specific dry weight ratios of production to residue, combustion efficiency and percentages of crop residues burning in field and domestic burning as fuel in China used in the emission calculations, as mentioned in Section 2. Summary table for the average annual mercury emission from biomass burning at provincial-level in China, as described in Section 3. This information is available free of charge via the Internet at http://pubs.acs.org.

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ACKNOWLEDGMENT

This study was funded by National Natural Science Foundation of China (No. 40975088, 70821140353, and 70773001) and the Public Welfare Projects for Environmental Protection (200809122).

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